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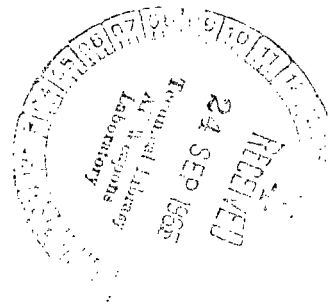


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BETHE-PEIERLS-WEISS APPROXIMATION AND A MODEL FOR FERROMAGNETIC THIN FILMS

by Gale Fair

*Lewis Research Center
Cleveland, Ohio*





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SUMMARY

The cluster approximation developed by P. R. Weiss to treat the problem of the Heisenberg ferromagnet is used to calculate the ferromagnetic properties of a two-layer rectangular lattice, a simple cubic lattice, and a body-centered cubic lattice, for spin $1/2$ atoms. The two-layer rectangular lattice is considered as a model for a simple cubic ferromagnetic thin film, and the results are compared with the properties of a complete simple cubic lattice. The results obtained indicate that such a two-layer lattice shows ferromagnetic behavior, with a Curie temperature about two-thirds that of the simple cubic lattice. The magnetization, internal field, energy, specific heat, and paramagnetic susceptibility are calculated and compared for the three lattices. One feature that appears in this calculation for the two-layer lattice, as previously obtained for the simple cubic case, is the occurrence of an "anti-Curie" temperature; a second, lower, temperature at which the spontaneous magnetization vanishes.

INTRODUCTION

The existence of ferromagnetism for some metals (e.g., iron, nickel, cobalt, and some of their alloys) is a phenomenon for which no completely adequate theoretical explanation has yet been given. The actual interaction mechanism that gives rise to ferromagnetism is not fully understood. In practice a Hamiltonian is assumed for a model of this interaction, such as the Heisenberg Hamiltonian, which will be used in this work. Once the Hamiltonian is specified, however, the problem is not finished. For example, in the case of the Heisenberg model, the Hamiltonian eigenvalue problem cannot be solved exactly for a crystal. To determine the thermodynamic properties of a system from this model, several techniques have been developed for making approximate calculations.

One of these approximation methods is the method of spin waves first introduced by Bloch (ref. 1). The original Bloch theory predicts the existence of a Curie temperature for a three-dimensional lattice but finds no such Curie temperature for a planar, two-dimensional lattice. This method has also been applied to the case of thin ferromagnetic films (refs. 2 and 3), to determine the effect of film thickness upon the spontaneous magnetization for films in which the thickness of the lattice is quite small in comparison with the planar dimensions. The results of spin-wave theory applied to finite lattices (refs. 2 and 3) indicate that such systems, at sufficiently low temperatures, show ferromagnetic behavior, even to the limit of lattices that are only one atomic layer in thickness.

The primary motivation of the present analysis was to determine whether these results of spin-wave theory were obtainable with other methods of dealing with ferromagnetic phenomena. The method selected was the Bethe-Peierls-Weiss approximation (or BPW method), which is basically a high-temperature approximation for treating the Heisenberg Hamiltonian. This method is applied herein to a new model for a ferromagnetic thin film. This thin-film model consists of two plane quadratic (square) lattices parallel in space with the distance between the planes equal to the interatomic distance within each lattice. This means that each atom will have five nearest neighbors.

In addition to the analysis of this thin-film model, a more complete investigation is made than was available of the properties of other simple lattices. This permits comparison of the results for the thin-film model, not only with the low temperature spin-wave calculations, but also with the properties of bulk samples treated with the BPW method.

To clarify the nature and source of the BPW method to be used, it is necessary to review briefly the more elementary Ising model, with its approximate methods of solution, as well as the Heisenberg model.

Ising Model

The Ising model represents a method devised by Ising (ref. 4) (and subsequently developed further), to treat a class of cooperative phenomena. This model has been used in the theory of ferromagnetism, the "lattice gas," order-disorder in binary alloys, and other problems involving interactions between atoms situated upon lattice sites. It is assumed that there is an interaction energy between a lattice atom and each of its nearest neighbors in the lattice. For example, in ferromagnetism this energy is to depend upon the relative orientation of the spins of the atoms; for order-disorder it depends upon whether the neighboring atoms are like or unlike species; and so forth. The problem is to use statistical mechanical techniques to calculate the thermodynamic properties of the system from a knowledge of its total energy.

As a model for ferromagnetism, the Ising model is too highly idealized to predict quantitative results for a real ferromagnet. At most, this model may be used as a guide for some of the qualitative aspects of a ferromagnetic system. In this vein, Onsager (ref. 5) has given a rigorous calculation for the two-dimensional Ising system and finds that such a lattice has a Curie temperature.

The total energy for the Ising model is determined by the Hamiltonian for the system. This Ising Hamiltonian has the following form:

$$\mathcal{H}_{\text{IS}} = -2 \sum_{i>j} \epsilon S_i S_j - B \sum_i S_i \quad (1)$$

The first summation represents a double sum, one over all spins S_i , and the second over all the nearest neighbors to S_i , denoted by S_j , where S_i represents, for ferromagnetism, the value of the scalar spin of the atom that is on the i^{th} lattice site, and ϵ is the interaction energy between S_i and S_j . The second term represents the interaction between the spin system and an external magnetic field proportional to B .

Among the approximate methods used to solve this problem, the one of particular interest here was developed by Bethe (ref. 6) for the solution of the order-disorder problem in binary alloys. It was later shown by Peierls (ref. 7) that this method also could be extended to the treatment of ferromagnetism. Basically, this approximation consists of treating rigorously the interaction of a single atom with its nearest neighbors. The remainder of the system is then replaced by an equivalent internal field that interacts only with this first shell of nearest neighbor atoms. This problem can be solved exactly and the thermodynamic properties determined from the corresponding partition function, which includes the internal field. Since there must be no difference between the properties of the atom chosen as central and those for an atom in the first shell, a self-consistency condition is used to eliminate the internal field. In practice, this is done by requiring that the average magnetic moment of the central atom in terms of the external field and the average magnetic moment of an atom in the first shell in terms of the internal field be calculated and set equal. This calculation explicitly determines the internal field.

This procedure may be extended to a series of approximations. At each step one more shell of atoms is included in the rigorous calculation, the remainder still being replaced by the internal field. This procedure is extremely difficult beyond the first step, however.

The method used in this report is an extension of the Bethe-Peierls approximation developed for the Ising model (as described previously), to the Heisenberg exchange Hamiltonian.

Heisenberg or Exchange Interaction Model

The Heisenberg model (ref. 8) for ferromagnetism was developed as a consequence of the exchange interaction of quantum mechanics. This exchange interaction between a pair of spins arises from the requirements of the Pauli exclusion principle and the Coulomb interaction and may be thought of as a result of the overlapping of the wave functions describing the atoms.

Similar in form to the Ising Hamiltonian, the Heisenberg Hamiltonian for a system of spins that occupy lattice sites is given by

$$\mathcal{H}_H = -2 \sum_{i>j} J \bar{S}_i \cdot \bar{S}_j - B \sum_i S_{iz} \quad (2)$$

plus an unimportant constant term involving only J . The summation is over nearest neighbor pairs, J is the exchange interaction coupling constant, and S_{iz} is the z -component of the i^{th} spin. The vector spins here are to be treated as quantum-mechanical operators.

Among the methods used to treat equation (2) is an extension due to P. R. Weiss (ref. 9) of the Bethe-Peierls approximation for the Ising model. This method (referred to as the Bethe-Peierls-Weiss approximation, or simply the BPW method) is the one used herein. The details of the calculation will be given in the next section of this report, but it is basically the Bethe-Peierls method applied to the exchange interaction. One atom is chosen as the central atom, and its interaction with the first shell of nearest neighbor atoms is calculated exactly; the rest of the system is replaced by an internal field that acts only upon this first shell. The corresponding energy eigenvalue problem can be solved and the resulting energy levels used to calculate the partition function for the system. From this partition function and the self-consistency condition on the average magnetic moments of the atoms, the ferromagnetic properties for the lattice may be determined.

THEORY

Energy Levels and Partition Function

The cluster Hamiltonian for the BPW approximation is obtained from the Heisenberg exchange Hamiltonian (eq. (2)), which is for the entire crystal, by the procedure sketched in the introduction. The exchange interaction between the spin of the chosen central atom \bar{S}_0 and the resultant spin of the first shell of nearest neighbors \bar{S}_1 is extracted from the

exchange term in equation (2). The remaining interactions of the first-shell atoms with their nearest neighbors and with the applied magnetic field are then written as an interaction with an effective internal field \bar{B}_1 . The external applied field \bar{B}_0 defines the z-axis of the system. In this approximation the internal field \bar{B}_1 , as described before, is assumed to be in the same direction. Aside from the exchange interactions, the spin of the central atom interacts only with the externally applied field, and only the first shell of atoms interacts with the internal field. With these conditions, the Hamiltonian for a cluster in which there are no interactions between atoms within the first shell has the following form:

$$\mathcal{H} = -2J\bar{S}_0 \cdot \bar{S}_1 - g\beta S_{0z}B_0 - g\beta S_{1z}B_1 \quad (3)$$

where

J	exchange integral
S_{0z}	z-component of spin of central atom
S_{1z}	z-component of resultant spin of first shell
B_0	magnitude of external field
B_1	magnitude of internal field
g	Lande spin factor
β	Bohr magneton

For convenience in notation, $g\beta B_0$ will be replaced by H_0 and $g\beta B_1$ by H_1 .

The eigenvalues of $\bar{S}_0 \cdot \bar{S}_1$ may be calculated from the vector model of angular momentum as follows:

$$\bar{S} = \bar{S}_0 + \bar{S}_1$$

Therefore,

$$\bar{S} \cdot \bar{S} = \bar{S}_0 \cdot \bar{S}_0 + \bar{S}_1 \cdot \bar{S}_1 + 2\bar{S}_0 \cdot \bar{S}_1$$

thus,

$$2\bar{S}_0 \cdot \bar{S}_1 = \bar{S} \cdot \bar{S} - \bar{S}_0 \cdot \bar{S}_0 - \bar{S}_1 \cdot \bar{S}_1 \quad (4)$$

Now define the quantity E_{ex} as the expectation value of the exchange interaction, as follows:

$$E_{\text{ex}} \equiv -2J \langle \bar{S}_0 \cdot \bar{S}_1 \rangle \quad (5)$$

Thus E_{ex} will have the values E_- and E_+ for $S = S_1 - \frac{1}{2}$ and $S = S_1 + \frac{1}{2}$, respectively. Then, from equation (4)

$$\left. \begin{aligned} E_- &= J(S_1 + 1) & S &= S_1 - \frac{1}{2} \\ E_+ &= -JS_1 & S &= S_1 + \frac{1}{2} \end{aligned} \right\} \quad (6)$$

when the spin per atom is $1/2$, the only case considered here.

The representation used is one in which the exchange energy is diagonal. Since $S_{0z} + S_{1z} = S_z = m$, the z-component of the total spin, by rearrangement of the field-dependent terms in equation (3),

$$S_{0z}H_0 + S_{1z}H_1 = S_{0z}H_0 + (m - S_{0z})H_1 = mH_1 - S_{0z}(H_1 - H_0)$$

In this representation, the energy levels when $m = \pm(S_1 + \frac{1}{2})$ are

$$W = -JS_1 \mp \frac{1}{2} (2S_1H_1 + H_0)$$

When $|m| \leq (S_1 - \frac{1}{2})$, the matrix for the Hamiltonian is (see appendix)

$$\begin{pmatrix} A_a & B \\ B & A_p \end{pmatrix}$$

where

$$\left. \begin{aligned} A_a &= J(S_1 + 1) - mH_1 - \frac{m}{2S_1 + 1} (H_1 - H_0) \\ A_p &= -JS_1 - mH_1 + \frac{m}{2S_1 + 1} (H_1 - H_0) \\ B &= \frac{1}{2} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right]^{1/2} (H_1 - H_0) \end{aligned} \right\} \quad (7)$$

For convenience, the following definitions are made:

$$x \equiv H_1 - H_0$$

$$\alpha \equiv \frac{m}{2S_1 + 1}$$

The determinantal equation for the eigenvalues then becomes

$$\begin{vmatrix} E_- - mH_1 - \alpha x - \lambda & \frac{1}{2} (1 - 4\alpha^2)^{1/2} x \\ \frac{1}{2} (1 - 4\alpha^2)^{1/2} x & E_+ - mH_1 + \alpha x - \lambda \end{vmatrix} = 0 \quad (8)$$

The resulting quadratic equation for λ_k , the eigenvalues, may be solved exactly and yields

$$\lambda_k = \frac{E_+ + E_-}{2} - mH_1 \pm \frac{E_+ - E_-}{2} [1 + \xi x(4\alpha + \xi x)]^{1/2} \quad (9)$$

where

$$\xi \equiv \frac{1}{E_+ - E_-} = - \frac{1}{J(2S_1 + 1)}$$

The term ξx is proportional to the ratio $(H_1 - H_0)/J$. Ultimately H_0 will be set equal to zero; in the neighborhood of the Curie point, the internal field H_1 is small, so that the square root may be expanded in terms of the ratio ξx . After expanding to fourth order and rearranging terms, the energy levels are

$$\lambda_k \equiv W_k = A_k + b_k H_1 + c_k (H_1 - H_0) + d_k (H_1 - H_0)^2 + e_k (H_1 - H_0)^3 + f_k (H_1 - H_0)^4 \quad k=1, 2 \quad (10)$$

where

$$A_{1,2} = E_{\mp}$$

$$b_1 = b_2 = -m$$

$$\begin{aligned}
c_{1,2} &= \mp \frac{m}{2S_1 + 1} \\
d_{1,2} &= \pm \frac{1}{4J(2S_1 + 1)} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right] \\
e_{1,2} &= \pm \frac{m}{2J^2(2S_1 + 1)^3} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right] \\
f_{1,2} &= \mp \frac{1}{16J^3(2S_1 + 1)^3} \left[1 - \frac{24m^2}{(2S_1 + 1)^2} + \frac{80m^4}{(2S_1 + 1)^4} \right]
\end{aligned}$$

Terms to fourth order in the field strengths are kept for the specific heat calculation although only the second-order terms are needed in explicit form for the determination of the Curie temperature.

The partition function Z for the cluster, written in terms of the energy levels W_k , then takes on the following form:

$$Z = \sum_{S_1} \omega(2n, S_1) \left[\sum_{m_-} e^{-W_1(S_1, m_-)/kT} + \sum_{m_+} e^{-W_2(S_1, m_+)/kT} \right] \quad (11)$$

The first summation is over the value of the total spin of the first shell S_1 from $S_1 = 0$ to $S_1 = n$, where $2n$ is the number of nearest neighbor atoms within the first shell. The summation indicated by m_- is over m from $m = -(S_1 - \frac{1}{2})$ to $m = +(S_1 - \frac{1}{2})$, and the m_+ sum is from $m = -(S_1 + \frac{1}{2})$ to $m = +(S_1 + \frac{1}{2})$. The factor $\omega(2n, S_1)$ represents the degeneracy of each S_1 level. This is essentially the number of ways that the first shell of atoms may have a total spin of S_1 . To calculate this factor, let $L(2n, S_1)$ stand for the number of ways of placing $2n$ spins, $n + S_1$ up and $n - S_1$ down, upon the $2n$ lattice sites, giving a z-component of the total spin equal to S_1 , with the total spin of S_1 or greater. Then

$$L(2n, S_1) = \frac{(2n)!}{(n + S_1)!(n - S_1)!} \quad (12)$$

The corresponding factor for the arrangement of $n + S_1 + 1$ up spins and $n - S_1 - 1$ down spins upon the $2n$ sites, resulting in a total spin of $S_1 + 1$ or greater, is $L(2n, S_1 + 1)$

$$L(2n, S_1 + 1) = \frac{(2n)!}{(n + S_1 + 1)!(n - S_1 - 1)!} \quad (13)$$

The difference between these two factors is just the number of arrangements upon the $2n$ sites that give a total spin exactly equal to S_1 , with a z-component also equal to S_1 . Thus

$$\begin{aligned} \omega(2n, S_1) &= L(2n, S_1) - L(2n, S_1 + 1) \\ &= \frac{(2n)!}{(n + S_1)!(n - S_1)!} - \frac{(2n)!}{(n + S_1 + 1)!(n - S_1 - 1)!} \end{aligned} \quad (14)$$

Self-Consistency Condition and Curie Temperature

The expression for the partition function (eq. (11)) still contains the internal field explicitly. To eliminate this from the calculation, the consistency condition must be applied to the system; that is, the central atom must be identical to any atom in the first shell. Explicitly, this means that the average magnetic moment of the central atom must be equal to the average magnetic moment of an atom in the first shell. Since the central atom does not interact with the internal field, the average magnetic moment \overline{m}_0 is given by

$$\overline{m}_0 = kT \frac{\partial \ln Z}{\partial B_0} = g\beta kT \frac{\partial \ln Z}{\partial H_0} \quad (15)$$

The average magnetic moment of a first-shell atom, which is determined by only the internal field, is given by $1/2n$ times the average magnetic moment of the entire first shell and is

$$\overline{m}_1 = \frac{1}{2n} kT \frac{\partial \ln Z}{\partial B_1} = \frac{1}{2n} g\beta kT \frac{\partial \ln Z}{\partial H_1} \quad (16)$$

Thus, for the consistency condition, \overline{m}_0 and \overline{m}_1 must be equal

$$2n \frac{\partial \ln Z}{\partial H_0} = \frac{\partial \ln Z}{\partial H_1} \quad (17)$$

The solution of this equation for the internal field can be obtained for the case of a vanishing external field. Thus, after the derivatives of the partition function are taken, H_0 is set equal to zero. In evaluating equation (17) odd functions of m must vanish, since the sums are taken over an interval symmetric about $m = 0$. Equation (17) gives the following equation determining the internal field explicitly:

$$AH_1 + CH_1^3 = 0 \quad (18)$$

which for nonvanishing H_1 yields

$$A + CH_1^2 = 0 \quad (19)$$

The quantities A and C are defined as

$$\left. \begin{aligned} A &\equiv \sum_{k=1}^2 \sum_{S_1=0}^n \omega(2n, S_1) \left(A_k^0 + \frac{A_k^1}{\ell T} \right) e^{-A_k/\ell T} \\ C &\equiv \sum_{k=1}^2 \sum_{S_1=0}^n \omega(2n, S_1) \left[C_k^0 + \frac{C_k^1}{\ell T} + \frac{C_k^2}{(\ell T)^2} + \frac{C_k^3}{(\ell T)^3} \right] e^{-A_k/\ell T} \end{aligned} \right\} \quad (20)$$

with

$$\left. \begin{aligned} A_k^0 &\equiv \sum_m (-2d_k) \\ (1 + 2n)A_k^1 &\equiv \sum_m \left[(b_k + c_k)^2 + 2nc_k(b_k + c_k) \right] \\ C_k^0 &\equiv \sum_m (-4f_k) \\ (1 + 2n)C_k^1 &\equiv \sum_m 2 \left[(1 + 2n)d_k^2 + (2 + 3n)b_k e_k + 2(1 + 2n)c_k e_k \right] \\ -(1 + 2n)C_k^2 &\equiv \sum_m \left[2d_k(b_k + c_k)^2 + 2nd_k(b_k + c_k)(b_k + 2c_k) \right] \\ 6(1 + 2n)C_k^3 &\equiv \sum_m \left[(b_k + c_k)^4 + 2nc_k(b_k + c_k)^3 \right] \end{aligned} \right\} \quad (21)$$

To evaluate these summations the following identities are used:

$$\left. \begin{aligned} \sum_{m=-T}^T 1 &= 2T + 1 \\ \sum_{m=-T}^T m^2 &= \frac{1}{3} T(T + 1)(2T + 1) \\ \sum_{m=-T}^T m^4 &= \frac{1}{15} T(T + 1)(2T + 1)(3T^2 + 3T - 1) \end{aligned} \right\} \quad (22)$$

It is important to note that in deriving equation (19) one factor of H_1 has been cancelled from each side. This implies that $H_1 = 0$ is always a solution to the consistency condition. This result is analogous to the Bethe-Peierls solution for the Ising problem, which also gives such a trivial solution. For a system for which there exists a nonzero Curie temperature, this solution will not be of interest. Physically, this zero solution corresponds to no long range order in the system.

The quantities A and C in equation (18), the consistency condition, are functions of the temperature, or, more precisely, functions of the ratio $x = J/kT$. Thus, H_1 is determined as a function of x or T . The Curie temperature T_c is specified by the vanishing of the internal field, or, equivalently, by the vanishing of the quantity A in equation (19). Explicitly this means evaluating A , setting the expression equal to zero, and solving for x . This equation has the general form

$$\sum_{S_1=0}^n \omega(2n, S_1) \frac{4}{3} \frac{S_1(S_1 + 1)}{(2S_1 + 1)^2} \left\{ \frac{2x}{1 + 2n} \left[\left(S_1 + \frac{1}{2} \right) \left(S_1 + \frac{3}{2} \right) (S_1 - n) e^{S_1 x} \right. \right. \\ \left. \left. + \left(S_1 - \frac{1}{2} \right) \left(S_1 + \frac{1}{2} \right) (1 + n + S_1) e^{-(S_1+1)x} \right] + \left[e^{S_1 x} - e^{-(S_1+1)x} \right] \right\} = 0 \quad (23)$$

Equation (23) contains the parameter n , which specifies the type of lattice under consideration. Thus equation (23) determines the Curie temperature for any lattice that has no interactions among the first-shell atoms. For the different values of n corresponding to the lattices of interest here, the implicit equations for $x_c = J/kT_c$ are as follows:

(1) For the linear chain $2n = 2$, equation (23) becomes

$$e^{3x} = 1 - \frac{3}{2}x \quad (24)$$

and the only real, nonnegative solution is $x_c = 0$. Since this is a solution that is not possible physically (infinite temperature), the only possible solution of equation (18) for this system is $H_1 = 0$.

(2) For the quadratic lattice $2n = 4$, equation (23) becomes

$$\frac{72}{25}e^{2x} - 5(8x - 1)e^x + (32x - 5)e^{-2x} + \frac{72}{5}\left(x - \frac{1}{5}\right)e^{-3x} = 0 \quad (25)$$

Again the only nonnegative, real solution is $x_c = 0$, making the only physically possible solution of equation (18) $H_1 = 0$. Thus, for this two-dimensional lattice there is no Curie temperature in agreement with the Bloch spin-wave theory.

(3) Similarly, for the simple cubic lattice $2n = 6$, equation (23) has the following form:

$$\begin{aligned} \frac{2}{49}(e^{3x} - e^{-4x}) + \frac{1}{5}(e^{2x} - e^{-3x}) + \frac{x}{7}\left(\frac{7}{2}e^{2x} - 9e^{-3x}\right) + \frac{1}{3}(e^x - e^{-2x}) \\ - \frac{5}{7}x\left(e^x - \frac{1}{2}e^{-2x}\right) + \frac{5}{7}xe^{-4x} = 0 \end{aligned} \quad (26)$$

One acceptable solution to this equation is $x_c = 0.540$ or $kT_c = 1.850 J$.

(4) For the model for a two-layer thin film, a lattice with five nearest neighbors, the equation for x_c is

$$\begin{aligned} \frac{7}{36}\left[4xe^{-3x} + \frac{1}{3}(e^{3x} - e^{-3x})\right] + \frac{1}{4}(e^{2x} - e^{-2x}) - \frac{x}{6}(3e^{2x} - 5e^{-2x}) + \frac{1}{4}(e^x - e^{-x}) - \frac{x}{3}e^x = 0 \end{aligned} \quad (27)$$

The solution of equation (27) is $x_c = 0.783$, corresponding to a Curie temperature given by $kT_c = 1.278 J$.

(5) The explicit form of equation (23) for the body-centered cubic lattice ($2n = 8$) is quite complicated, but the numerical solution gives the Curie temperature of $kT_c = 2.908$ J.

The previous results show a qualitative agreement with the Bloch spin-wave theory. The one- and two-dimensional lattices show no ferromagnetic behavior, that is, no Curie temperature. The three-dimensional space lattices have solutions for the Curie temperatures, while the five-nearest-neighbor lattice structure, constructed as an intermediate model, also has a Curie temperature, which falls below that for the simple cubic lattice.

Anti-Curie Temperature

In addition to the Curie temperature solutions for the consistency condition as discussed previously, there also appears a second set of solutions to equation (23) for those lattices for which there exists a nonzero solution for x . These second solutions, which occur at lower temperatures than T_c , have been termed anti-Curie points by P. W. Anderson (ref. 10), who first noticed their existence. For the lattices for which there is no real solution to equation (23), the question does not arise. The simple cubic lattice $2n = 6$ has such an anti-Curie point with the value $kT_{AC} = 0.269$ J as found by Anderson. For the two-layer model $2n = 5$ and for the body-centered cubic $2n = 8$, the second solutions occur at $kT_{AC} = 0.339$ J and $kT_{AC} = 0.212$ J, respectively.

The existence of a second zero of the internal field, and correspondingly, the vanishing of the magnetization of the system below T_{AC} is clearly in disagreement with the physical behavior of a ferromagnetic system.

The source of this difficulty can be traced to the expansion of the energy eigenvalues in powers of $(H_1 - H_0)/J$. The region of temperature for which this expansion is expected to be valid can be obtained by evaluating the internal field, obtained from equation (19), as a function of temperature. For the simple cubic lattice and also for the body-centered cubic $2n = 8$, there is a singularity in the internal field; the factor C in equation (19) vanishes. Numerical evaluation shows that this singularity occurs very near the Curie temperature, at a temperature of about 90 percent of the Curie temperature. For the two-layer lattice the factor C does not vanish, but it becomes small enough that the internal field becomes very large, also quite near the Curie temperature. The appearance of the anti-Curie point cannot be taken too seriously since in all cases it occurs outside what must be considered as the region of validity of the energy eigenvalue expansion.

Figures 1 and 2 show the calculated dimensionless internal field and magnetization \overline{m}_0 very near the critical temperature for the three lattices under consideration. All curves show the extremely rapid decrease to zero at the transition temperature, which is characteristic of ferromagnetic transitions.

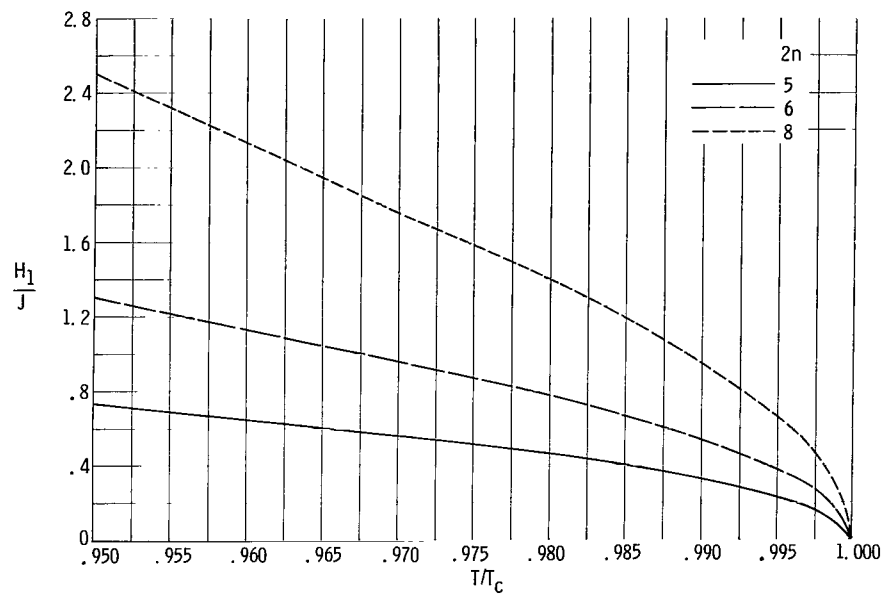


Figure 1. - Internal field as function of reduced temperature.

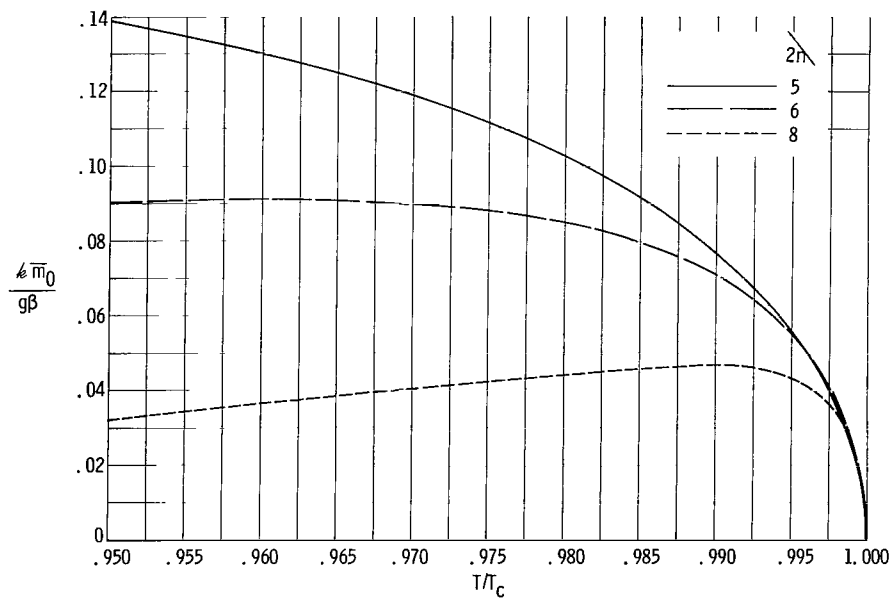


Figure 2. - Magnetization as function of reduced temperature.

Energy and Specific Heat

Since the specific heat of the system is the temperature derivative of the energy, the starting point for this calculation is the specification of the energy per atom of the system as a function of the temperature. The exchange term in equation (3) represents the interaction between a central atom and its shell of first nearest neighbor atoms. In a system with N atoms there are $\frac{1}{2}N$ such interactions. The energy of the entire system may be written as $\frac{1}{2}N$ times the ensemble average of the exchange energy

$$E = \frac{1}{2} N \left(-2J \langle \bar{S}_0 \cdot \bar{S}_1 \rangle \right) \quad (28)$$

where the angular brackets indicate ensemble averages and $H_0 = 0$. Now, since $2\bar{S}_0 \cdot \bar{S}_1 = S(S+1) - S_1(S_1+1) - S_0(S_0+1)$ with S_0 , S_1 , and S defined as before and since $S_0 = 1/2$, then

$$2\langle \bar{S}_0 \cdot \bar{S}_1 \rangle = \left\langle S(S+1) - S_1(S_1+1) - \frac{3}{4} \right\rangle$$

Thus, taking the ensemble average of the exchange term

$$2\langle \bar{S}_0 \cdot \bar{S}_1 \rangle = Z^{-1} \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m \left[S(S+1) - S_1(S_1+1) - \frac{3}{4} \right] e^{-W_k/kT}$$

where

$$Z \equiv \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m e^{-W_k/kT}$$

results in the energy of the system having the form

$$E = \frac{1}{2} N Z^{-1} \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m A_k e^{-W_k/kT} \quad (29)$$

with all symbols as defined previously.

As with the discussion following equation (19), the summand in the region near the Curie temperature may be expanded and the energy per atom \mathcal{E} may be obtained

$$\mathcal{E} \equiv \frac{E}{N} = \frac{1}{2} Z^{-1} \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m A_k e^{-A_k/kT} \left\{ \left(1 - \frac{b_k + c_k}{kT} \right) H_1 + \left[\frac{1}{2} \frac{(b_k + c_k)^2}{(kT)^2} - \frac{d_k}{kT} \right] H_1^2 \right\}$$

or

$$\begin{aligned} \mathcal{E} = \frac{1}{2} Z^{-1} \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m A_k e^{-A_k/kT} \\ + \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m A_k e^{-A_k/kT} \left[\frac{1}{2} \frac{(b_k + c_k)^2}{(kT)^2} - \frac{d_k}{kT} \right] H_1^2 \end{aligned}$$

For convenience in computation the energy may be rewritten in terms of four new functions of the temperature to second order in H_1 as

$$\frac{\mathcal{E}}{J} = \frac{1}{2} \frac{P_1}{P_0} + \frac{1}{2} H_1^2 \left(\frac{P_3}{P_0} - \frac{P_1 P_2}{P_0^2} \right) \quad (30)$$

where Z has been rewritten as

$$Z = P_0 + P_2 H_1^2$$

and P_0 , P_1 , P_2 , and P_3 are defined as

$$P_0 \equiv \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m e^{-A_k/kT} \quad (31)$$

$$P_1 \equiv \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m A_k e^{-A_k/kT} \quad (32)$$

$$P_2 \equiv \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m \left[\frac{1}{2} \frac{(b_k + c_k)^2}{(kT)^2} - \frac{d_k}{kT} \right] e^{-A_k/kT} \quad (33)$$

$$P_3 \equiv \sum_{S_1} \omega(2n, S_1) \sum_k \sum_m A_k \left[\frac{1}{2} \frac{(b_k + c_k)^2}{(kT)^2} - \frac{d_k}{kT} \right] e^{-A_k/kT} \quad (34)$$

A further solution requires an explicit evaluation of these functions for the specific lattice of interest, that is, $2n = 5, 6$, or 8 .

In addition to the temperature dependence, the energy equation (eq. (30)) contains the internal field explicitly. This may be eliminated by use of the self-consistency condition (eq. (19)) giving

$$H_1^2 = -\frac{A}{C}$$

From these results the energy per atom has been calculated numerically as a function of the reduced temperature as displayed in figure 3 for the two-layer lattice, the simple cubic lattice, and the body-centered cubic lattice, very near the Curie temperature. It is evident, both from the expression for the energy (eq. (30)) and from the curves, that the energy is a continuous function of the reduced temperature but with a discontinuity in

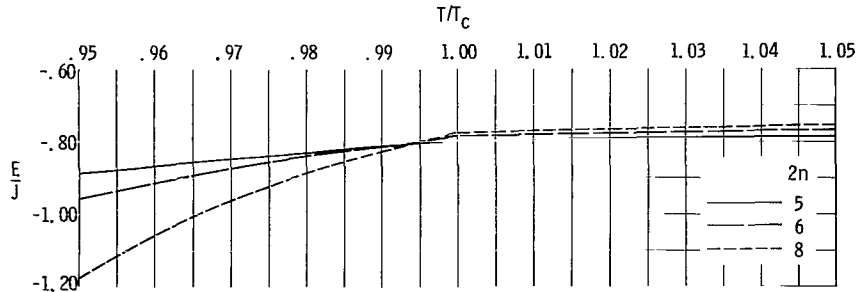


Figure 3. - Energy as function of reduced temperature.

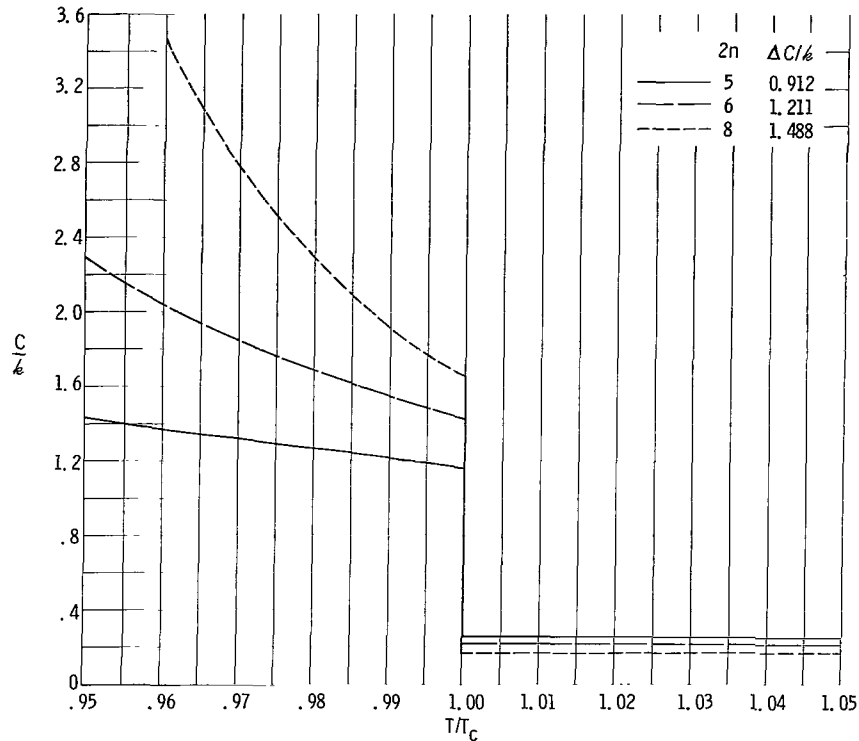


Figure 4. - Specific heat as function of reduced temperature.

slope at the transition temperature.

Since the specific heat of the system is simply the temperature derivative of the energy, there is a finite discontinuity in the specific heat at the Curie temperature, as displayed in figure 4. The specific heat discontinuity implies that this system undergoes a second-order phase transition, according to the Ehrenfest definition, at the Curie temperature. The order parameter in the system is the internal field, just as in the original Weiss molecular field formulation. The values of the discontinuities are 0.912, 1.211, and 1.488 for the two-layered lattice, the simple cubic lattice, and the body-centered cubic lattice, respectively.

Paramagnetic Susceptibility

As is readily seen from the graph of the spontaneous magnetization for temperatures above the Curie temperature, the internal field vanishes in the absence of an applied field. When there is an applied field present, the situation is not the same. The applied field interacting with the spins produces an internal field and a resulting magnetization. This is experimentally verified by the observation that above the Curie temperature there is a paramagnetic susceptibility which obeys a modified Curie-Weiss law of the form

$$\chi = \frac{C}{T - \theta} \quad (35)$$

where C is a constant, the Curie constant, and θ is called the paramagnetic Curie temperature. Typically, θ is found to be a few percent larger than T_c , the ferromagnetic Curie temperature.

The paramagnetic susceptibility may be calculated from the BPW formalism by starting with the partition function given by equation (11), but keeping a nonzero applied field. For an expansion very near the Curie temperature only terms that are second order in the field strengths need be kept. Just as before, the average magnetic moment of the central atom in the cluster and the average magnetic moment of an atom in the first shell of nearest neighbors may be calculated from equations (15) and (16). The self-consistency condition requires that these be equal, giving now an equation between the applied field H_0 and the internal field H_1

$$AH + BH_0 = 0 \quad (36)$$

where H is the resultant internal field, $H = H_1 - H_0$, A is given by equation (20), and B has the form

$$B = \sum_{k=1}^2 \sum_{S_1=0}^n \omega(2n, S_1) \sum_m \left[\frac{b_k^2 + (1 + 2n)b_k c_k}{(1 + 2n)kT} \right] e^{-A_k/kT} \quad (37)$$

In the calculation of the susceptibility, it is more convenient to work with the average magnetic moment of the entire cluster \bar{m}

$$\bar{m} = \bar{m}_0 + 2n\bar{m}_1$$

from which the magnetization \bar{m}_0 is

$$\bar{m}_0 = \frac{\bar{m}}{1 + 2n} \quad (38)$$

after the self-consistency condition has been applied.

$$\frac{kT}{g\beta} \frac{\bar{m}}{H_0} = \sum_{S_1} \omega(2n, S_1) \sum_k \sum_{m_k} \left[b_k^2 + b_k(b_k + c_k) \frac{H}{H_0} \right] e^{-A_k/kT} \quad (39)$$

The susceptibility then is the ratio of the magnetization \bar{m}_0 to the applied magnetic field B_0 (or $H_0/g\beta$). Thus from equations (39) and (38)

$$\chi = \frac{g\beta\bar{m}_0}{H_0} = \frac{g^2\beta^2}{kT} \frac{1}{1+2n} \sum_{S_1} \omega(2n, S_1) \sum_k \sum_{m_k} \left[b_k^2 + b_k(b_k + c_k) \frac{H}{H_0} \right] e^{-A_k/kT} \quad (40)$$

The applied field H_0 and the internal field H enter only as a ratio, and this may be eliminated by use of the consistency equation (36). Equation (40) may be written as

$$\frac{kT}{g^2\beta^2} \chi = \frac{Z^{-1}}{1+2n} \left[\sum_{S_1} \omega(2n, S_1) \sum_k \sum_{m_k} b_k^2 e^{-A_k/kT} - \frac{B}{A} \sum_{S_1} \omega(2n, S_1) \sum_k \sum_{m_k} b_k(b_k + c_k) e^{-A_k/kT} \right] \quad (41)$$

For a comparison with the Curie-Weiss law (eq. (35)), it is more convenient to calculate the reciprocal of the susceptibility. This curve then should be linear with an intercept on the T -axis that is equal to the paramagnetic Curie temperature θ . If equation (41) is rewritten to correspond with this discussion,

$$\frac{g^2\beta^2}{kT} \frac{1}{\chi} = (1+2n)Z \left(\chi_0 - \frac{B}{A} \chi_H \right)^{-1} \quad (42)$$

where χ_0 and χ_H are the corresponding summations in equation (41). Results have

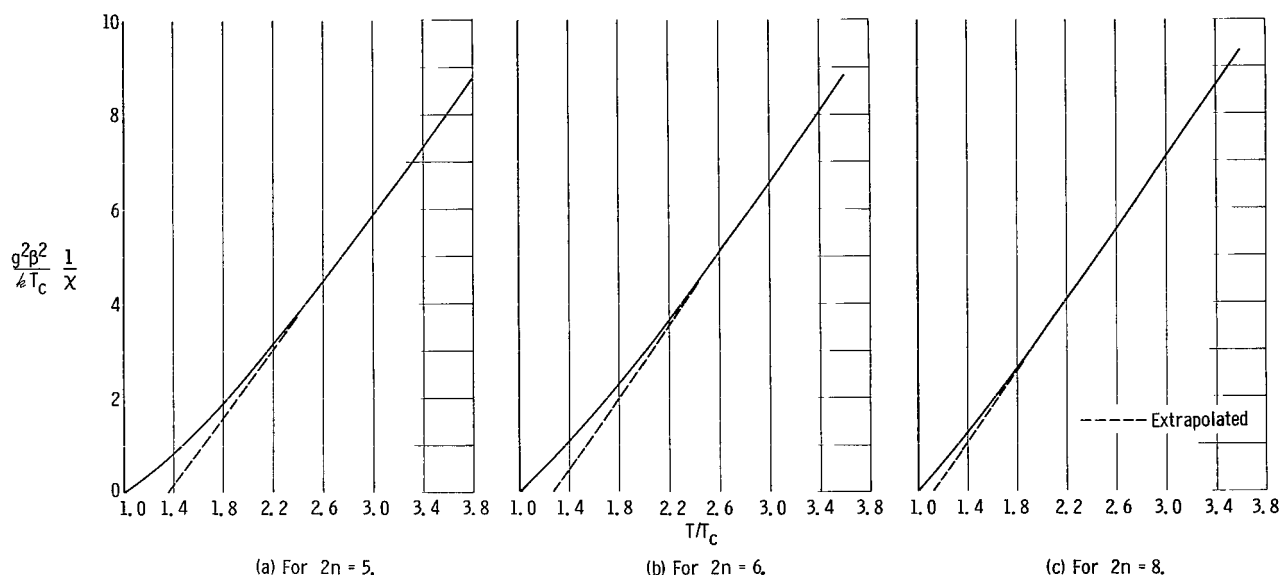


Figure 5. - Reciprocal of paramagnetic susceptibility as function of reduced temperature.

been calculated for $2n = 5, 6$, and 8 , and are plotted in figures 5(a), (b), and (c), respectively.

A numerical investigation of the various terms in equation (42) shows that the zero of the reciprocal susceptibility occurs when the factor A vanishes. But this point has already been found to define the ferromagnetic Curie temperature. Thus, what could be defined as the rigorous paramagnetic Curie temperature coincides with the ferromagnetic Curie temperature, at least in the cases considered here with the BPW approximation. Figure 5 shows, however, that another paramagnetic Curie temperature can be defined by extrapolating the linear portion of the curves down to the axis. The resulting values for the Curie temperature obtained by this scheme are 1.36, 1.26, and 1.12 times the ferromagnetic Curie temperature for the two-layer lattice, the simple cubic lattice, and the body-centered cubic lattice, respectively. These extrapolated numbers do not have much significance due to the remaining nonlinearity in the curves for large T/T_c (as seen in fig. 5).

DISCUSSION

To fulfill the original purpose for this study, it is appropriate at this point to make a comparison with the spin-wave results of Bloch (ref. 1), which predict a ferromagnetic Curie temperature for three-dimensional lattices, but none for two-dimensional systems. The results of the calculations presented here, applicable only to infinite lattices, seem to essentially agree with the results of Bloch. The linear chain and plane quadratic lattices show no ferromagnetic behavior, while the three-dimensional lattices with five, six,

and eight nearest neighbors have solutions for the Curie temperature. The Curie temperature for the two-layer thin-film model occurs at approximately two-thirds the value of the Curie temperature for a simple cubic lattice, both calculated with the BPW approximation.

The correspondence between the results of the two methods is not as complete as it first appears. The requirement for a lattice to have ferromagnetic properties as a result of the BPW method is of a more topological nature than the corresponding criterion for the Bloch theory. According to the BPW method, if there are no interactions between the first-shell atoms, any lattice that has a coordination number (number of nearest neighbors) of more than four has a Curie temperature. A calculation by Weiss (ref. 9), shows that lattices with interactions between the atoms in the first shell must have a coordination number greater than or equal to eight (e. g. , a modified body-centered cubic) to possess ferromagnetic behavior.

As a point of contrast between the Bloch and BPW methods consider the two-layer model under consideration in this report, that is, a coordination number of five. It has been shown that such a lattice has a Curie temperature, as indicated by the nonzero solution to equation (27). Although this represents a three-dimensional lattice, it is of interest to consider an imaginary two-dimensional lattice having the same coordination number. The fact that such a lattice does not satisfy the symmetry requirements for a two-dimensional Bravais lattice is not important in this discussion. If such a plane lattice with coordination number five could be constructed, it would have a Curie temperature identical to that calculated for the two-layer lattice, that is, it would also satisfy equation (27).

From this example and the discussion earlier, it appears that the requirement for a lattice to be ferromagnetic on the BPW approximation may be stated in a concise form: For a lattice of a particular class (i. e. , presence or absence of interactions among the first-shell atoms) to possess ferromagnetic properties it is necessary and sufficient that the lattice coordination number be equal to or greater than some minimum value (e. g. , $2n \geq 5$ for noninteracting lattices).

One point for comparison of the BPW approximation with a real ferromagnetic system arises from equation (30). From this equation it is seen that the energy per atom of such a system is a continuous function of the temperature with a finite jump discontinuity in the slope at the Curie temperature. There is some evidence, on the other hand, to indicate that there should actually be a logarithmic singularity in the slope (for a discussion see Hill (ref. 11)). As yet, however, the experiments have not been precise enough to settle definitely the question of the nature of the singularity at the ferromagnetic Curie point.

It is of interest to compare the results of this calculation using the BPW approximation with the "constant-coupling" approximation of P. W. Kastelijn (ref. 12). This

method assumes that the Heisenberg Hamiltonian for the entire system may be written in terms of effective scalar two-body interaction Hamiltonians. From these two-body terms the energy of the system may be calculated by means of standard thermodynamic techniques.

The interest in this method arises from the fact that for a cluster that contains no interactions between first-shell atoms, the BPW method represents, in a sense, a system of two-body interactions, just as in the Kastelijn approximation. This correspondence is not complete, since the BPW method includes long-range correlations inherent in the assumption about the internal field, not included in the constant-coupling method. For the two-layer lattice, however, first-shell interactions are absent, and the constant-coupling method gives a useful cross-check for the calculations done previously.

The expressions derived by Kastelijn for the Curie temperature T_c and the discontinuity in the specific heat ΔC are

$$k T_c = 2J \left(\ln \frac{2n}{2n-4} \right)^{-1}$$

$$\Delta C = \frac{3}{32} \frac{4n^2(2n-4)^2}{(2n-1)(2n-2)} \left(\ln \frac{2n}{2n-4} \right)^2 k$$

where $2n$ represents the number of nearest neighbors for the lattice, as used before. Evaluating these quantities for $2n = 5$ results in

$$T_c = 1.243 \frac{J}{k}$$

$$\Delta C = 0.506 k$$

which compare fairly well with our value of $1.278 \frac{J}{k}$ for the Curie temperature, but not with the specific heat discontinuity ($0.912 k$) obtained from the BPW approximation.

There are two points to be noted in making a comparison between the BPW and the constant-coupling approximations. First, the condition that the lattice must have $2n \geq 5$ for ferromagnetic behavior to exist is the same in each method. Secondly, in the constant-coupling approximation there is no second solution corresponding to the anti-Curie point as appears in the BPW calculations.

CONCLUDING REMARKS

Within the limitations of the Bethe-Peierls-Weiss approximation the two-layer lattice

model for a thin film shows behavior that is qualitatively consistent with the results of spin-wave theory and also with the properties of noninteracting bulk lattices as calculated by the BPW method. The Curie temperature is consistent with that calculated for the simple cubic and body-centered cubic, and the second-order nature of the phase transition appears as a finite jump discontinuity in the specific heat.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, June 1, 1965.

APPENDIX - CLUSTER HAMILTONIAN

This appendix deals with the problem of calculating the matrix elements of the Hamiltonian (eq. (3)) in a representation in which the exchange energy is diagonal. The discussion essentially follows that of Condon and Shortley (ref. 13). The Hamiltonian may be written in terms of H_0 and H_1 as discussed in the text

$$\mathcal{H} = -2J\bar{S}_0 \cdot \bar{S}_1 - H_0 S_{0z} - H_1 S_{1z} \quad (A1)$$

where the vector sum of \bar{S}_0 and \bar{S}_1 is defined by

$$\bar{S} = \bar{S}_0 + \bar{S}_1 \quad (A2a)$$

and

$$S_z = S_{0z} + S_{1z} \quad (A2b)$$

Then, by means of equations (4) and (A2b) \mathcal{H} may be rewritten in a more useful form

$$\mathcal{H} = -J(\bar{S}^2 - \bar{S}_0^2 - \bar{S}_1^2) - S_z H_1 + S_{0z}(H_1 - H_0) \quad (A3)$$

The representation that diagonalizes the exchange interaction is the representation in which \bar{S}^2 , S_z , \bar{S}_0^2 , and \bar{S}_1^2 are diagonal. The eigenvalues of the exchange energy are given by equation (6) and the eigenvalues of S_z are just the values of m . All that remains is to determine the matrix elements of S_{0z} in this representation.

To accomplish this, consider the properties of a class of operators known as "class T" operators. These operators are defined by their commutation relations with any angular momentum operator \bar{J} . These relations are

$$\left. \begin{array}{lll} [J_x, T_x] = 0 & [J_y, T_x] = -iT_z & [J_z, T_x] = iT_y \\ [J_x, T_y] = iT_z & [J_y, T_y] = 0 & [J_z, T_y] = -iT_x \\ [J_x, T_z] = -iT_y & [J_y, T_z] = iT_x & [J_z, T_z] = 0 \end{array} \right\} \quad (A4)$$

and

$$[\bar{J}, \bar{T}_1 \cdot \bar{T}_2] = 0 \quad (A5)$$

where square brackets symbolize commutator brackets and $\hbar = 1$. Any vector operator that transforms in the same manner as the position vector under a proper coordinate rotation is a class \mathbf{T} operator. In particular, an angular momentum operator, such as \mathbf{S}_0 , belongs to this class of operators.

To calculate the matrix elements of \mathbf{S}_{0z} , plan on taking the matrix elements of one component of a vector operator \mathbf{T} . For this purpose, consider

$$\begin{aligned} [\bar{\mathbf{J}}^2, \bar{\mathbf{T}}] &= \bar{\mathbf{J}} \cdot [\bar{\mathbf{J}}, \bar{\mathbf{T}}] - [\bar{\mathbf{T}}, \bar{\mathbf{J}}] \cdot \bar{\mathbf{J}} \\ &= -i(\bar{\mathbf{J}} \times \bar{\mathbf{T}} - \bar{\mathbf{T}} \times \bar{\mathbf{J}}) \\ &= -2i(\bar{\mathbf{J}} \times \bar{\mathbf{T}} - i\bar{\mathbf{T}}) \end{aligned}$$

which follow from the commutation relations. Now consider the following commutator:

$$\begin{aligned} [\bar{\mathbf{J}}^2, [\bar{\mathbf{J}}^2, \bar{\mathbf{T}}]] &= -2i[\bar{\mathbf{J}}^2, \bar{\mathbf{J}} \times \bar{\mathbf{T}} - i\bar{\mathbf{T}}] \\ &= -2i[\bar{\mathbf{J}}^2, \bar{\mathbf{J}} \times \bar{\mathbf{T}}] - 2[\bar{\mathbf{J}}^2, \bar{\mathbf{T}}] \\ &= 2(\bar{\mathbf{J}}^2 \bar{\mathbf{T}}^2 + \bar{\mathbf{T}} \bar{\mathbf{J}}^2) - 4\bar{\mathbf{J}}(\bar{\mathbf{J}} \cdot \bar{\mathbf{T}}) \end{aligned}$$

But there is also another expression for $[\bar{\mathbf{J}}^2, [\bar{\mathbf{J}}^2, \bar{\mathbf{T}}]]$

$$[\bar{\mathbf{J}}^2, [\bar{\mathbf{J}}^2, \bar{\mathbf{T}}]] = \bar{\mathbf{J}}^4 \bar{\mathbf{T}} - 2\bar{\mathbf{J}}^2 \bar{\mathbf{T}} \bar{\mathbf{J}}^2 + \bar{\mathbf{T}} \bar{\mathbf{J}}^4$$

Therefore,

$$\bar{\mathbf{J}}^4 \bar{\mathbf{T}} - 2\bar{\mathbf{J}}^2 \bar{\mathbf{T}} \bar{\mathbf{J}}^2 + \bar{\mathbf{T}} \bar{\mathbf{J}}^4 = 2(\bar{\mathbf{J}}^2 \bar{\mathbf{T}} + \bar{\mathbf{T}} \bar{\mathbf{J}}^2) - 4\bar{\mathbf{J}}(\bar{\mathbf{J}} \cdot \bar{\mathbf{T}}) \quad (\text{A6})$$

From this identity the matrix elements of $\bar{\mathbf{T}}$ can be calculated since $\bar{\mathbf{J}}$ is a Hermitian operator. For example, from the left side of equation (A6), for the matrix elements between the states $\langle j m |$ and $| j' m' \rangle$, as

$$\begin{aligned} \langle j m | \bar{\mathbf{J}}^4 \bar{\mathbf{T}} - 2\bar{\mathbf{J}}^2 \bar{\mathbf{T}} \bar{\mathbf{J}}^2 + \bar{\mathbf{T}} \bar{\mathbf{J}}^4 | j' m' \rangle &= \langle j m | \bar{\mathbf{J}}^4 \bar{\mathbf{T}} | j' m' \rangle - 2\langle j m | \bar{\mathbf{J}}^2 \bar{\mathbf{T}} \bar{\mathbf{J}}^2 | j' m' \rangle \\ &+ \langle j m | \bar{\mathbf{T}} \bar{\mathbf{J}}^4 | j' m' \rangle = [j(j+1) - j'(j'+1)]^2 \langle j m | \bar{\mathbf{T}} | j' m' \rangle \quad (\text{A7}) \end{aligned}$$

since a system of units is used in which $\hbar = 1$. Similarly for the right side of equation (A6),

$$\begin{aligned} \langle jm | 2(\bar{J}^2 \bar{T} + \bar{T} J^2) - 4\bar{J}(\bar{J} \cdot \bar{T}) | j'm' \rangle &= 2[j(j+1) + j'(j'+1)] \langle jm | \bar{T} | j'm' \rangle \\ &\quad - 4 \langle jm | \bar{J}(\bar{J} \cdot \bar{T}) | j'm' \rangle \end{aligned} \quad (A8)$$

But the right side of equation (A8) may be rewritten as

$$\langle jm | \bar{J}(\bar{J} \cdot \bar{T}) | j'm' \rangle = \sum_{j''m''} \langle jm | \bar{J} | j''m'' \rangle \langle j''m'' | \bar{J} \cdot \bar{T} | j'm' \rangle$$

But $\bar{J} \cdot \bar{T}$ is diagonal with respect to j and m , which follows from equation (A5); thus

$$\langle jm | \bar{J}(\bar{J} \cdot \bar{T}) | j'm' \rangle = \langle jm | \bar{J} | jm' \rangle \langle jm' | \bar{J} \cdot \bar{T} | jm' \rangle$$

Thus,

$$\begin{aligned} \left\{ [j(j+1) - j'(j'+1)]^2 - 2[j(j+1) + j'(j'+1)] \right\} \langle jm | \bar{T} | j'm' \rangle \\ = -4 \langle jm | \bar{J} | jm' \rangle \langle jm' | \bar{J} \cdot \bar{T} | jm' \rangle \end{aligned} \quad (A9)$$

This is the expression determining the matrix elements of \bar{T} , and in particular, those of T_z .

Note in equation (A9) that, if $j' = j \pm 1$, the square bracket on the left side vanishes. Looking only for the expression when $j = j'$ gives

$$j(j+1) \langle jm | \bar{T} | jm' \rangle = \langle jm | \bar{J} | jm' \rangle \langle jm' | \bar{J} \cdot \bar{T} | jm' \rangle \quad (A10)$$

Since J_z is diagonal in m , it follows for the z -component that

$$j(j+1) \langle jm | T_z | jm \rangle = \langle jm | J_z | jm \rangle \langle jm | \bar{J} \cdot \bar{T} | jm \rangle \quad (A11)$$

This expression determines the diagonal elements of S_{0z} in this representation.

Since equation (A9) is indeterminate for the off-diagonal elements, another expression must be derived that is valid when $j' = j \pm 1$. To do this, consider the ladder operators T_+ and T_- defined on the set of class T operators

$$\left. \begin{aligned} T_+ &= T_x + iT_y \\ T_- &= T_x - iT_y \end{aligned} \right\} \quad (A12)$$

Similar ladder operators may be defined for the \bar{J} operators, as

$$\left. \begin{aligned} J_+ &= J_x + iJ_y \\ J_- &= J_x - iJ_y \end{aligned} \right\} \quad (A13)$$

To express T_z in terms of these operators, consider the following commutator:

$$\begin{aligned} [J_+, T_-] &= [J_x + iJ_y, T_x - iT_y] \\ &= [J_-, T_-] + 2i[J_y, T_-] \end{aligned}$$

or, from equation (A4),

$$T_z = \frac{1}{2} [J_+, T_-] \quad (A14)$$

Thus, from this expression and a knowledge of the matrix elements of the ladder operators the matrix elements of T_z may be calculated. For example,

$$\langle jm | T_z | j'm \rangle = \frac{1}{2} \sum_{j''m''} \left(\langle jm | J_+ | j''m'' \rangle \langle j''m'' | T_- | j'm \rangle - \langle jm | T_- | j''m'' \rangle \langle j''m'' | J_+ | j'm \rangle \right) \quad (A15)$$

The ladder operators act only upon the m values and have nonzero matrix elements only between two states whose m -values differ by unity.

Consider each case in equation (A15) separately (see refs. 13 to 15)

(1) For $j' = j - 1$

$$\begin{aligned} 2\langle jm | T_z | j-1, m \rangle &= \langle jm | J_+ | j, m-1 \rangle \langle j, m-1 | T_- | j-1, m \rangle \\ &\quad - \langle jm | T_- | j-1, m+1 \rangle \langle j-1, m+1 | J_+ | j-1, m \rangle \\ &= \langle j: T: j-1 \rangle \sqrt{(j-m+1)(j-m)} \sqrt{(j+m)(j-m+1)} \\ &\quad - \langle j: T: j-1 \rangle \sqrt{(j-m)(j-m-1)} \sqrt{(j+m)(j-m+1)} \\ &= 2\sqrt{j^2 - m^2} \langle j: T: j-1 \rangle \end{aligned} \quad (A16)$$

(2) For $j' = j + 1$

$$\begin{aligned}
2\langle jm | T_z | j+1, m \rangle &= \langle jm | J_+ | j, m-1 \rangle \langle j, m-1 | T_- | j+1, m \rangle \\
&\quad - \langle jm | T_- | j+1, m+1 \rangle \langle j+1, m+1 | J_+ | j+1, m \rangle \\
&= 2\sqrt{(j+1)^2 - m^2} \langle j:T:j+1 \rangle
\end{aligned} \tag{A17}$$

where the quantities $\langle j:T:j-1 \rangle$ and $\langle j:T:j+1 \rangle$ are in the notation of Condon and Shortley (ref. 13) and need to be defined. Starting with the commutation relation for J_- and T_- , that is, $[J_-, T_-] = 0$, and calculating the matrix elements that satisfy the selection rules on m result in

$$\langle j, m-1 | J_- | jm \rangle \langle jm | T_- | j', m+1 \rangle = \langle j, m-1 | T_- | j'm \rangle \langle j'm | J_- | j', m+1 \rangle \tag{A18}$$

for $j' - j = 0, \pm 1$.

But the matrix elements of the J_- operator are well-known and yield

$$\sqrt{(j+m)(j-m+1)} \langle jm | T_- | j', m+1 \rangle = \sqrt{(j'+m+1)(j'-m)} \langle j, m-1 | T_- | j'm \rangle$$

Thus

$$\frac{\langle jm | T_- | j', m+1 \rangle}{\sqrt{(j'+m+1)(j'-m)}} = \frac{\langle j, m-1 | T_- | j', m \rangle}{\sqrt{(j+m)(j-m+1)}} \quad j' - j = 0, \pm 1 \tag{A19}$$

This relation between these ratios is independent of m .

This equality evaluated for $j' = j \pm 1$ is used to define the terms $\langle j:T:j-1 \rangle$ and $\langle j:T:j+1 \rangle$

$$\left. \begin{aligned}
\frac{\langle jm | T_- | j-1, m+1 \rangle}{\sqrt{(j-m-1)(j-m)}} &= \frac{\langle j, m-1 | T_- | j-1, m \rangle}{\sqrt{(j-m)(j-m+1)}} \equiv \langle j:T:j-1 \rangle \\
\frac{\langle jm | T_- | j+1, m+1 \rangle}{\sqrt{(j+m+2)(j+m+1)}} &= \frac{\langle j, m-1 | T_- | j+1, m \rangle}{\sqrt{(j+m+1)(j+m)}} \equiv -\langle j:T:j+1 \rangle
\end{aligned} \right\} \tag{A20}$$

These elements are related to the set of irreducible elements of a class \overline{T} operator. The evaluation of these will be reserved until later.

Equations (A11), (A16), and (A17) determine the matrix elements of S_{0z} explicitly, once a change of notation is made. Before this is done, it would be useful to consider the general form of the operator S_{0z} . This matrix will have $2\left(S_1 + \frac{1}{2}\right) + 1 = 2(S_1 + 1)$ rows and columns. The only nonzero elements will lie upon the principal diagonal and along the diagonals one step removed from the principal diagonal. This follows from the fact that the only nonzero matrix elements for \bar{T} are for $j' - j = \pm 1$. Each row or column is indexed by a value of S_1 , or, equivalently by m . Calculating the matrix elements of S_{0z} as given in the text results in effectively picking out a square 2 by 2 matrix with $j = S_1 + \frac{1}{2}$, $S_1 - \frac{1}{2}$ and for a given value of m . Thus to calculate the matrix elements from equation (A11), (A16), and (A17), the association is made that the j -values for the two states are $S_1 + \frac{1}{2}$ and $S_1 - \frac{1}{2}$, associating with T_z the operator S_{0z} , which is a class T operator.

First, for the diagonal elements of S_{0z} , $\langle jm | \bar{J} \cdot \bar{T} | jm \rangle$ is needed for $\bar{T} \rightarrow \bar{S}_0$ and $\bar{J} \rightarrow \bar{S}$. Consider $\bar{S}_1 = \bar{S} - \bar{S}_0$; then

$$\bar{S}_1^2 = \bar{S}^2 + \bar{S}_0^2 - 2\bar{S}_0 \cdot \bar{S}$$

or,

$$\bar{S}_0 \cdot \bar{S} = \frac{1}{2} (\bar{S}^2 + \bar{S}_0^2 - \bar{S}_1^2)$$

and then,

$$\left\langle S_1 + \frac{1}{2}, m | \bar{S}_0 \cdot \bar{S}_1 | S_1 + \frac{1}{2}, m \right\rangle = \frac{1}{2} \left[\left(S_1 + \frac{1}{2} \right) \left(S_1 + \frac{3}{2} \right) + \frac{3}{4} - S_1(S_1 + 1) \right] = \frac{1}{2} \left(S_1 + \frac{3}{2} \right)$$

and

$$\left\langle S_1 - \frac{1}{2}, m | \bar{S}_0 \cdot \bar{S}_1 | S_1 - \frac{1}{2}, m \right\rangle = -\frac{1}{2} \left(S_1 - \frac{1}{2} \right)$$

Thus,

$$\begin{aligned} \left\langle S_1 + \frac{1}{2}, m | S_{0z} | S_1 + \frac{1}{2}, m \right\rangle &= \frac{m}{\left(S_1 + \frac{1}{2} \right) \left(S_1 + \frac{3}{2} \right)} \frac{1}{2} \left(S_1 + \frac{3}{2} \right) \\ &= \frac{m}{2S_1 + 1} \end{aligned} \tag{A21}$$

$$\begin{aligned}
\left\langle S_1 - \frac{1}{2}, m | S_{0z} | S_1 - \frac{1}{2}, m \right\rangle &= \frac{m}{\left(S_1 - \frac{1}{2}\right)\left(S_1 + \frac{1}{2}\right)} - \frac{1}{2} \left(S_1 - \frac{1}{2}\right) \\
&= -\frac{m}{2S_1 + 1}
\end{aligned} \tag{A22}$$

For the off-diagonal matrix elements the irreducible terms appearing in equations (A16) and (A17) must be evaluated. When \bar{S}_0 is the operator \bar{T} involved, the evaluation of these terms may be derived, but the calculation is prohibitively long to be shown here. The details are given in Condon and Shortley (ref. 13, chapter 3); the results are merely quoted here

$$\left\langle S_1 + \frac{1}{2} : S_0 : S_1 + \frac{1}{2} \right\rangle = \left[\frac{(S - S_0 + S_1)(S + S_0 - S_1)(S + S_0 + S_1 + 1)(S_0 + S_1 + 1 - S)}{4S^2(2S - 1)(2S + 1)} \right]^{1/2}$$

for $S = S_1 + \frac{1}{2}$ and

$$\left\langle S_1 - \frac{1}{2} : S_0 : S_1 + \frac{1}{2} \right\rangle = \left[\frac{(S + 1 - S_0 + S_1)(S + 1 + S_0 - S_1)(S_0 + S_1 - S)(S_0 + S_1 + S + 2)}{\{4(S + 1)^2[4(S + 1)^2 - 1]\}} \right]^{1/2}$$

for $S = S_1 - \frac{1}{2}$.

Evaluating these gives

$$\left\langle S_1 + \frac{1}{2} : S_0 : S_1 - \frac{1}{2} \right\rangle = \frac{1}{2S_1 + 1} \tag{A23}$$

and

$$\left\langle S_1 - \frac{1}{2} : S_0 : S_1 + \frac{1}{2} \right\rangle = \frac{1}{2S_1 + 1} \tag{A24}$$

Substituting these results into equations (A16) and (A17), the matrix elements of S_{0z} are

$$\left\langle S_1 + \frac{1}{2}, m \mid S_{0z} \mid S_1 - \frac{1}{2}, m \right\rangle = \left\langle S_1 - \frac{1}{2}, m \mid S_{0z} \mid S_1 + \frac{1}{2}, m \right\rangle = \frac{1}{2} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right]^{1/2}$$

Thus, from equations (A21), (A22), and (A25), the matrix form of one of the 2 by 2 block matrices of S_{0z} is

$$S_{0z} = \begin{pmatrix} \frac{m}{2S_1 + 1} & \frac{1}{2} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right]^{1/2} \\ \frac{1}{2} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right]^{1/2} & -\frac{m}{2S_1 + 1} \end{pmatrix} \quad (\text{A26})$$

Accordingly, the matrix of the Hamiltonian is

$$\mathcal{H} = \begin{pmatrix} J(S_1 + 1) - mH - \frac{m}{2S_1 + 1} (H_1 - H_0) & \frac{1}{2} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right]^{1/2} (H_1 - H_0) \\ \frac{1}{2} \left[1 - \frac{4m^2}{(2S_1 + 1)^2} \right]^{1/2} (H_1 - H_0) & -JS_1 - mH_1 + \frac{m}{2S_1 + 1} (H_1 - H_0) \end{pmatrix} \quad (\text{A27})$$

as given in the text. These are the results given by Weiss (ref. 9).

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